

On page 1, after the title, please insert the following section heading.

BACKGROUND OF THE INVENTION

On page 1, please amend the section heading "Technical Field" to read as follows:

1. Field of the Invention

On page 1, please amend the section heading "Background of the Invention" to read as follows:

2. Description of the Related Art

On page 1, please delete the third complete paragraph and insert the following replacement paragraph:

High molecular weight products produced by hardening (or polymerizing) the above-mentioned compounds have various uses, especially epoxy resin and silicone resin which have been widely used as adhesives, sealing, and paints in various fields such as automobile industry, housing/building material industry, civil engineering and construction industries, aircraft industry, and electric and electronic industries.

On page 1, please delete the fifth complete paragraph and insert the following replacement paragraph:

It has been well known that the polymerization is generally carried out at room temperature or under heating conditions. Recently, searches for a photopolymerization catalyst or initiator have been conducted, because it may be preferable to carry out the polymerization under irradiation conditions depending upon use. However, no remarkable result has been obtained.

**On page 2, please delete the fourth complete paragraph and insert the following replacement paragraph:**

A<sup>3</sup>  
As another example, the photopolymerization initiator disclosed in Japanese Patent Publication No. 62692/94 of the same applicant as this application can be cited. This photopolymerization initiator comprises a composite obtainable from a reaction between (a) charge transfer complex consisting of biscyclopentadienyl iron derivative and quinoid and (b) at least one salt selected from a group consisting of tetrafluoroborates, hexafluorophosphates and hexafluoroantimonates.

09800438 073104  
A<sup>4</sup>  
**On page 2, please delete the fifth complete paragraph and insert the following replacement paragraph:**

On the contrary, regarding the photopolymerization of organosiloxane, it is very hard to find an example. An example can be found only in Kevin D. Belfield et al., "Photoinitiated cationic ring-opening polymerization of a cyclosiloxane", Polymer Bulletin 38, pp. 165-168 (1997). Belfield et al. disclosed that when hexamethylene cyclotrisiloxane is exposed with U.V. light in the presence of sulfonium or iodonium salt or iminosulfonate derivative, it is subjected to a photo-initiating cationic ring-opening polymerization in solution or in the absence of solvent.

**Please delete the paragraph bridging pages 2 and 3 and insert the following replacement paragraph:**

A<sup>5</sup>  
Regarding the thermal hardening (polymerization) reaction which is carried out at room temperature or under heating conditions, as example of catalyst for polymerization of epoxy compound, tertiary amine (such as benzylmethyl amine, 2,4,6-trisdimethylamine methylphenol, etc.), imidazole (such as 2-methylimidazole, 2-ethyl-4-methylimidazole, 2-heptadecylimidazole etc.), Lewis acid (such as BF<sub>3</sub> monoethylamine, BF<sub>3</sub> piperazine, etc.) have been known.

**On page 3, please delete the first complete paragraph and insert the following replacement paragraph:**

A<sup>6</sup>  
The thermal hardening reaction of organosiloxane compound is classified into addition type and condensation type: in a reaction of the former type, peroxide or platinum compound is used as catalyst and in a reaction of the latter type, metal salt of carboxylic acid is used.

**On page 3, please delete the second complete paragraph and insert the following replacement paragraph:**

A<sup>2</sup>  
As mentioned above, epoxy resins and silicone resins have been widely used. However, on producing the epoxy resins or silicone resins by hardening (or polymerizing) each monomer, the catalyst or initiator used in the thermal polymerization or photopolymerization of epoxy compound is completely different from that used in the thermal polymerization or photopolymerization of organosiloxane compound, and additionally, an initiator for photopolymerization of organosiloxane compound is almost unknown.

**On page 3, please amend the section heading "Disclosure of the Invention" to read as follows:**

Summary of the Invention

**On page 4, please amend the section heading "Best Mode for Carrying out the Invention" to read as follows:**

Description of the Preferred Embodiments

**On page 4, please delete the third complete paragraph which begins with "The crystalline" and insert the following replacement paragraph:**

A<sup>8</sup>  
The crystalline ion-association substance of the present invention comprises, as is seen from the general formula (I), a metallocene derivative cation and a tetradentate borate complex anion wherein the four ligands are the same to each other.

**Please delete the paragraph bridging pages 4 and 5 and insert the following replacement paragraph:**

A<sup>9</sup>  
In the general formula (I), the electron donative or electron attractive substituent is alkyl group, cycloalkyl group, aryl group, alkoxy group, silyl group, dialkyl group, acyl group, cycloalkenyl group, amino group, carboxyl group, organoboranyl group, phosphino group, aldehyde group, hydroxyl group and the like. More particularly, the alkyl group is selected from among lower alkyl groups such as methyl group, ethyl group, propyl group and butyl group, or pentyl group or amyl group; and the cycloalkyl group is selected from among cyclobutyl group, cyclopentyl group, cycloheptyl group, and cyclohexyl group. The aryl group is selected from among phenyl group, naphtyl group, etc.

**On page 6, please delete the paragraph which begins with "On the other hand,".**

**Before the paragraph bridging pages 6 and 7, please insert the following paragraph:**

A<sup>10</sup>  
On the other hand, the counter anion of the crystalline ion-association substance of the present invention is tetradentate borate complex anion  $[B(R^2)_4]^-$ . In the formula,  $R^2$  is a ligand coordinated to boron atom (B) of the center nucleus and is selected from among aryl group, halogenated aryl group, halogen haloformaryl group, cycloalkynyl group, halogenated cycloalkyl group, halogenated cycloalkynyl group, cycloalkyloxy group, cycloalkenyloxy group, alkadienyl group, alkatrienyl group, alkynyl group, halogenated

ENP A<sub>10</sub>  
alkenyl group, halogenated alkadienyl group, halogenated alkatrienyl group, halogenated alkynyl group, heterocyclic group, etc., but the four ligands ( $R^2$ ) are the same to each other. Also, the two adjacent ligands may chemically bond to each other to form two rings bonding the ligands within one borate complex anion.

**On page 8, please delete the first complete paragraph and insert the following replacement paragraph:**

A"  
036043-073104  
The crystalline ion-association substance of the present invention contains a bulky ligand in the cyclopentadienyl moiety of the metallocene derivative cation. Therefore, a transition metal of the center nucleus having a high oxidation state may maintain thermal stability, and the crystallinity between the metallocene derivative cation and the anion complex to be associated is synergistically increased.

**On page 11, please delete the first complete paragraph and insert the following replacement paragraph:**

A  
The crystalline ion-association substance of the present invention may have such a structure that one of four identical ligands of the borate complex anion is closed to the transition metal center of the metallocene derivative cation and is sandwiched between the two cyclopentadiene ligands of the metallocene derivative cation; the metallocene derivative cation per se having such a structure that the two cyclopentadiene ligands with various structure are positioned in the form of a dihedral structure with respect to the transition metal center.

**Please delete the paragraphs bridging pages 11 and 12 and insert the following replacement paragraph:**

A<sup>13</sup>  
In photopolymerization of a cationically polymerizable organic substance, the polymerization initiator of the present invention is used in the amount of from 0.1 to 10 weight parts, preferably 0.5 to 4 weight parts with respect to 100 weight parts of the

EN<sup>13</sup> A  
cationically polymerizable organic substance; when the cationically polymerizable organic substance absorbs usually 2000 to 9000 mJ/cm<sup>2</sup> of energy by irradiation with U.V. light having the wavelength of 200-700nm, basically 200-400nm, a high molecular weight polymer is produced. However, an irradiation operation with more energy may be required depending upon the type of polymerizable substance. The irradiation can be carried out at any condition, that is, at ordinary temperature, under cooling condition, or under heating condition; and at atmospheric pressure or vacuum, or in the presence of inert gas.

On page 12, please delete the third complete paragraph and insert the following replacement paragraph:

A  
138504383073404  
When compatibility between the cationically polymerizable organic substance and the polymerization initiator is poor, a suitable solvent such as hydrocarbon solvents, halogenated hydrocarbon solvents, alcoholic solvents, phenolic solvents, ether/acetal solvents, ketone solvents, ester solvents, nitrogen-containing compound solvents may be used. The concrete examples of the solvent are dichloromethane, chloroform, methanol, tetrahydrofuran, acetone, methylethyl ketone, acetonitrile, etc. The solvent is used in 5-3 times amount with respect to 100 weight parts of the cationically polymerizable organic substance, that is, 500-300 weight parts.

On page 17, please delete subparagraph III) (b) and insert the following replacement paragraph:

A<sup>15</sup>  
(b) the hardened product has the substantially unchanged characteristics of the starting material itself; for example, when it is mixed with the cationically polymerizable organic substance such as epoxy compound, the obtainable polymer product (hardened product) has the unchanged inherent characteristics of epoxy compound, and additionally, it is possible to produce composite product (hardened product) consisting of at least two cationically polymerizable organic substances which are of a different type from each other.

**On page 18, please delete the second paragraph and insert the following replacement paragraph:**

A<sup>16</sup>  
In preparation of the crystalline ion-association substance of the present invention, commercially available products (for example, products sold by Tokyo Chemicals Co. or Aldrich Co.) can be used as metallocene derivative. Other metallocene derivatives can be prepared by the well-known methods for preparation disclosed in the following references:

**On page 22, please delete the first complete paragraph and insert the following replacement paragraph:**

0380438-073304  
A  
By using a similar apparatus and procedure to those used in Example 4, 0.46g of (dihydroxyboryl) ferrocenium/tetrakis(3,5-difluorophenyl) borate was obtained (yield 51%), starting from 0.3g of commercially available ferroceneboronic acid (CAS 12152-94-2), 0.021g of iron(III) chloride, 30ml of acetonitrile, 10ml of pure water and 0.635g of tetrakis (3,5-difluorophenyl) borate sodium salt.

**On page 23, please delete the second complete paragraph beginning with "By using".**

**On page 23, after the section heading under "Example 8" please insert the following new paragraph:**

A<sup>18</sup>  
By using similar apparatus and procedure to those used in the Example 1, 1.36g of dimethyl ferrocenium/tetrakis[3,5-bis(trifluoromethyl)phenyl] borate was obtained (yield 54.1%), starting from 0.5g of dimethyl ferrocene, 10g of conc. Sulfuric acid, and ethanol solution (7ml) containing 2.15g of tetrakis[3,5-bis(trifluoromethyl)phenyl]borate sodium salt.

Please delete the paragraph bridging pages 26 and 27 and insert the following replacement paragraph:

On the other hand, in case of using an organosiloxane compound having volatility and subliming property, a test sample was prepared as follows:

A<sup>19</sup>  
In a similar bottle to that described above, 10 weight parts of dichloromethane is added with respect to 100 weight parts of the organosiloxane compound, and then the predetermined amount of polymerization initiator is added thereto. The resulting liquid mixture is stirred to disperse the initiator. Thereafter, the liquid mixture enclosed in the bottle is irradiated repeatedly with U.V. light ( $6000\text{mJ/cm}^2$ ) four times from outside of the bottle. The obtained test sample (rubber-like hardened product produced in the bottle) is taken out and transferred to a glass cylinder for Soxhlet extraction test (glass filter with filter plate No. GP-100) to measure gel fraction.

On page 32, please delete the second complete paragraph and insert the following replacement paragraph:

A<sup>20</sup>  
To 100 weight parts of decamethylcyclopentasiloxane (D5) or dodecamethylcyclohexasiloxane (D6), 2 weight parts of each polymerization initiator which comprises the crystalline ion-association substance prepared in the Example 8 or 10, 4 weight parts of benzophenone (sensitizer) and 30 weight parts of dichloromethane were added. The obtained mixture was stirred and mixed to prepare a solution in which the organosiloxane compound, the sensitizer and the polymerization initiator were homogeneously dissolved in the solvent. The obtained solution was fed to the center recess of the hole slide glass of one hole type without overflowing the solution from the recess. The hole slide glass was put into a hot air circulating oven and maintained at  $60^\circ\text{C}$  for 10 min. The test film was cooled to room temperature and irradiated with U.V. light ( $600\text{ mJ/cm}^2$ ). The gel fraction of the hardened films was reported in the following Table 4.



On page 33, please delete the second complete paragraph and insert the following replacement paragraph:

21  
A  
00000438-073104  
To 100 weight parts of 1,3-bis(glycidoxypopyl) tetramethyldisiloxane (silicone-modified epoxy monomer) (Gelest Inc.; SIH1115.0; CAS 126-80-7), 1 weight part of each polymerization initiator which comprises the crystalline ion-association substance prepared in the Example 2, 3, 8, 9, 10, 11, 12 or 13, 2 weight parts of benzophenone (sensitizer) and 50 weight parts of dichloromethane were added. The obtained mixture was stirred and mixed to prepare a solution in which the silicone-modified epoxy compound, the sensitizer and the polymerization initiator were homogeneously dissolved in the solvent. The obtained solution was fed to the center recess of the hole slide glass of one hole type without overflowing the solution from the recess. The hole slide glass was put into a hot air circulating oven and maintained at 60°C for 10 min. and then at 80°C for 3 min. Each test film was irradiated with U.V. light (8000mJ/cm<sup>2</sup>).

On page 40, please delete the first complete paragraph and insert the following replacement paragraph:

22  
A  
These examples show the fact that the polymerization initiator which comprises the crystalline ion-association substance prepared in the above-mentioned Example 3, 9, 11 or 12 has an ability to produce a thermal polymerization of a mixture consisting of cyclic siloxane and epoxy compound.

On page 42, please delete the first complete paragraph beginning with "The polymerization" and insert the following replacement paragraph:

23  
A  
The polymerization initiator of the present invention has the unique property that it can initiate both photopolymerization and thermal polymerization when used in polymerizing cationically polymerizable organic substance, though the polymerization initiator comprises an identical crystalline ion-association substance.

**On page 42, please delete the second complete paragraph beginning with "In particular" and insert the following replacement paragraph:**

A<sup>24</sup> In particular, since the photopolymerization initiator for organosiloxane has been little developed, the polymerization initiator of the present invention is valuable.

**Please delete the paragraph bridging pages 42 and 43 and insert the following replacement paragraph:**

A<sup>25</sup> For the above-mentioned unique property of the polymerization initiator of the present invention, it is possible to obtain a merit in practical use; that is, after the U.V. light irradiation, it is possible to carry out the hardening in a shade zone which is not exposed to U.V. light or a deep zone to which U.V. light does not reach by means of heating (photopolymerization plus thermal polymerization). This merit is very important, because, in accordance with conventional method, when the hardening is carried out by using both means of photopolymerization and thermal polymerization in accordance with conventional method, it is necessary to modify a monomer itself; for example, to prepare a modified organosiloxane having epoxy group.

**On page 43, please delete the last complete paragraph and insert the following replacement paragraph:**

A<sup>26</sup> Therefor, it is possible to extend the field and method for use of cationically polymerizable organic substances by using the polymerization initiator of the present invention.